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The University of Michigan

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November 4, 1980

Dr. Howard E. Schlossberg, Program Manager
Directorate of Physics
AFOSR/NP
Bolling Air Force Base
Washington, DC 20332

Dear Dr. Schlossberg:

FINAL REPORT

AFOSR Grant-77-3225

Resonant and Non-Resonant Optical Frequency Mixing in Simple Molecular Systems

March 1, 1977-August 31, 1980

1) Molecular Electric Tensor Properties and Nonlinear Optics

(See Journal Articles 1, 2, 3, 4, 5, 6, 9, 12)

The motivation here is to pursue a deeper understanding of nonlinear optics by studying it in the simplest possible media together with the complementary goal of an improved understanding of molecular physics by intercomparison of experimental results, *ab initio* theory and simple models.

We have enjoyed an ongoing discussion with groups of quantum chemists (notably Bartlett, et al at Battelle, Arrighini, et al and Lazzeretti, et al) undertaking state-of-the-art *ab initio* computations of molecular coefficients. A number of aggravating differences between conventions have been eliminated and our rigorous and precise experimental data has made it clear that only the most elaborate computations are adequate and then only at the 20-50% level for several-atom molecules.

Journal article 3 provides experimental data for some relatively large molecules with delocalized electrons for comparison with a contemporary approximate computation. We conclude that such computations for molecules of this size are inadequate.

In collaboration with experimentalists using the Kerr effect to extract molecular nonlinear coefficients (Orr, Buckingham, Bogaard, Dunmur, Richie among others) we have investigated intriguing discrepancies between the results of their technique and ours. One possible source of these discrepancies (suggested by B. J. Orr) is from terms in a perturbation expression for a nonlinear coefficient involving excited vibrational states. The contributions are currently not included in even the most sophisticated computation. Journal article 12 is an experimental approach to this problem.

Since the success of *ab initio* theories seems not to be commensurate with the effort involved, we have investigated simple models of molecular nonlinear response such as the Bond Additivity Approximation and a model where bond-bond interactions are included (Journal articles 1, 2, and 6). Article 6 in particular has clarified the effectiveness and limitations of such models.

Anisotropic linear molecular polarizabilities are of interest in the context of molecular models and journal article 5 presents experimental data from a novel method involving intensity dependent refractive index in gases.

2) Resonant Nonlinear Optical Interactions

(See Journal Articles 7, 10, 11)

The presence of resonances offers the possibility of substantial enhancement of nonlinear optical coefficients. However, resonant enhancement is typically accompanied by a plethora of deleterious or complicating processes and may be severely limited by saturation. We have investigated resonant processes in cesium vapor in considerable detail.

THESES PRESENTED

A. V. Smith	"Three Resonant Nonlinear Optical Processes in Atomic Cesium". 1977.
C. K. Miller	"Nonlinear Optical Polarizabilities of Molecules". 1978.

SEMINARS GIVEN

"Measurement and Correlation of Hyperpolarizabilities for a Number of Halogenated Methanes"
C. K. Miller
Seminar presented at Sandia Laboratories
September, 1977

"Three Resonant Nonlinear Optical Processes in Atomic Cesium"
A. V. Smith
Seminar presented at Sandia Laboratories
March, 1978

"Three Resonant Nonlinear Optical Processes in Atomic Cesium"
A. V. Smith
Seminar presented at Joint Institute of Laboratory Astrophysics
July 1978

"Three Resonant Nonlinear Optical Processes in Atomic Cesium"
A. V. Smith
Seminar presented at Bell Labs, Murray Hill
June 1978

"Three Resonant Nonlinear Optical Processes in Atomic Cesium"
A. V. Smith
Seminar presented at Western Electric
June 1978

"Nonlinear Electric Polarizability of Delocalized Electrons"
J. F. Ward
Seminar presented at Wayne State University
November, 1978

"Resonant Nonlinear Processes in Cesium Vapor"
J. F. Ward
Seminar presented at Sandia Laboratories
December, 1978

"Nonlinear Optics and Molecular Hyperpolarizabilities"
J. F. Ward
Seminar presented at Battelle Institute
March, 1979

"Nonlinear Optics and Molecular Hyperpolarizabilities"
J. F. Ward
Seminar presented at University of Melbourne, Australia
July, 1979

"Nonlinear Optics and Molecular Hyperpolarizabilities"
J. F. Ward
Seminar presented at the University of New South Wales, Australia
July, 1979

"Collisional Processes and Two-Photon-Resonant Phase Conjugation in Cesium Vapor"
M. T. Myers
Seminar presented at Shell Research Laboratories, Houston
February, 1980

"Polarizability Anisotropies from Measurements of Intensity-Dependent Refractive Index in Gases"
D. S. Elliott
Seminar presented at Aerospace Corporation
September, 1980

"Polarizability Anisotropies from Measurements of Intensity-Dependent Refractive Index in Gases"
D. S. Elliott
Seminar presented at the University of Waterloo
September, 1980

"Polarizability Anisotropies from Measurements of Intensity-Dependent Refractive Index in Gases"
D. S. Elliott
Seminar presented at the Joint Institute of Laboratory Astrophysics
September, 1980

JOURNAL ARTICLES

1. "Measurements of Nonlinear Optical Polarizabilities for Some Halogenated Methanes: The Role of Bond-Bond Interactions", C. K. Miller and J. F. Ward, Phys. Rev. A16, 1179 (1977).

2. "An Interacting Segment Model of Molecular Electric Tensor Properties: Theory and Application to Electric Dipole Moments of the Halogenated Methanes", C. K. Miller, B. J. Orr, and J. F. Ward, J. Chem. Phys. 67, 2109 (1977).

3. "Measurement of Molecular Hyperpolarizabilities for Ethylene, Butadiene, Hexatriene, and Benzene", J. F. Ward and D. S. Elliott, J. Chem. Phys. 69, 5438 (1978).

4. "Measurements of Nonlinear Optical Polarizabilities for Twelve Small Molecules", J. F. Ward and C. K. Miller, Phys. Rev. A19, 826 (1979).

5. "Polarizability Anisotropies of CO_2, N_2O and OCS from Measurements of Intensity-Dependent Refractive Index in Gases", D. S. Elliott and J. F. Ward, (Submitted to Phys. Rev. Letters).

6. "An Interacting Segment Model of Molecular Electric Tensor Properties: Application to Dipole Moments, Polarizabilities, and Hyperpolarizabilities for the Halogenated Methanes", C. K. Miller, B. J. Orr, and J. F. Ward (Submitted to J. Chem. Phys.).

7. "Doubly-Resonant Four-Photon Interactions in Cesium Vapor", A. V. Smith and J. F. Ward (Submitted to IEEE J. of Quantum Electronics).

8. "Propagation Aspects of Nonlinear Optical Interactions Involving Intersecting Focused Beams", J. W. Dudley II and J. F. Ward (Submitted to Applied Optics).

9. "Measurements of Third-Harmonic Wavevector Mismatch Using a Two-Wavelength, Single-Path Interferometer", D. S. Elliott and J. F. Ward (Planned).

10. "Cesium Spin Exchange Cross Sections", M. T. Myers and J. F. Ward (Planned).

11. "Two-Photon Resonant Phase Conjugation in Cesium Vapor", M. T. Myers and J. F. Ward (Planned).

12. "Vibrational Contributions to Molecular Hyperpolarizabilities", D. S. Elliott and J. F. Ward (Planned).

PARTICIPANTS

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A. V. Smith	Graduate Student	1972-1977
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Respectfully submitted,

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